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Elastic stiffness and ultrasonic attenuation of superconductor MgB_2 at low temperatures

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Temperature dependencies of elastic constants and ultrasonic attenuation of a polycrystalline MgB_2 showing high-T_c superconductivity are reported. An electromagnetic acoustic resonance method detected the specimen's resonance frequencies, from which we derived the elastic constants for nonporous MgB_2 using micromechanics calculation. The bulk and shear moduli extrapolated to 0 K are determined to be 96.6 GPa and 74.2 GPa, respectively, which gives the Debye temperature $\Theta_D = 819$ K and the coupling constant $\lambda \approx 0.76$–0.89 with McMillan formula. In the temperature behavior of ultrasonic attenuation, two anomalous peaks are observed near 30 K.

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The remarkable discovery of intermetallic superconductor MgB_2 with a high T_c of 39 K (Ref. 1) has triggered related extensive studies to elucidate the mechanism of its superconductivity. A significant boron isotope effect[2] implies that MgB_2 is a phonon-mediated superconductor. The *ab initio* calculations,[3,4] tunneling-spectroscopy studies,[5] nuclear spin-lattice relaxation,[6] and photoemission measurements[7] have concluded that MgB_2 is a conventional *s*-wave superconductor of ''strong'' or ''intermediate'' electron-phonon coupling, although an essential uncertainty still remains in whether MgB_2 is a *multiple-gap*[8–11] or *anisotropic-gap*[12,13] superconductor.

The elastic constants are important parameters for knowing the Debye temperature and the electron-phonon coupling constant in a phonon-mediated superconductor. Several related studies appeared: first-principles calculations predicted the single-crystal elastic constants;[14,15] x-ray measurements under high pressure gave the *isothermal* bulk modulus;[16] the flexural resonant method gave Young's modulus;[17] and other results were given in sound-velocity measurements.[18] However, these are inconsistent with each other. Also the temperature dependence has not been systematically measured yet.

Here we have examined the elastic and anelastic properties of MgB_2 through the ultrasonic characteristics. This paper presents the temperature dependence of elastic constants of nonporous MgB_2 deduced using a micromechanics model, the temperature behavior of ultrasonic attenuation, and observation of two anomalous attenuation peaks below 30 K.

A sample was prepared from commercially available powder by pseudo-HIP sintering at 1273 K for 12 h (referring to the method described in Ref. 1), and we checked that T_c of the sample was about 39 K by an eddy-current method. The x-ray-diffraction spectrum indicated that the sample included a very small amount of MgO. The sample was machined into a rectangular parallelepiped, measuring $5.125(x_1) \times 3.940(x_2) \times 2.349(x_3)$ mm^3, where x_i denotes the coordinate axis of the specimen. The mass density is 2301 kg/m^3, 87.7% of the theoretical value.

The resonance frequencies for all vibration modes (dilatation, torsion, shear, and flexure modes) were measured by the resonant ultrasound spectroscopy (RUS) method[19] at room temperature. Subsequently, each resonance frequency was identified with the electromagnetic acoustic resonance (EMAR) method, which can excite the specific vibration group through the Lorentz-force mechanism. This noncontact technique[20] is quite sensitive to the ultrasonic attenuation and can measure accurately the internal (intrinsic) friction. Figure 1 shows the resonance spectrum, the resonance peak of the B_{2g}-2 vibration mode (-2 denotes the order of the resonance), and the free-decay amplitude.

At low temperatures, we used only the EMAR measurement and focused on the two vibration groups: B_{2g} (shear modes around the x_2 axis) and A_g (dilatation modes). The ultrasonic attenuation was measured by the free-decay method, and the attenuation coefficient α was determined by fitting $\exp(-\alpha t)$ to the measured ring-down with time. The normalized attenuation coefficient (internal friction) is obtained by $Q^{-1} = \alpha / \pi f$.

In the RUS procedure, the elastic constants are determined by comparing the measured and calculated resonance

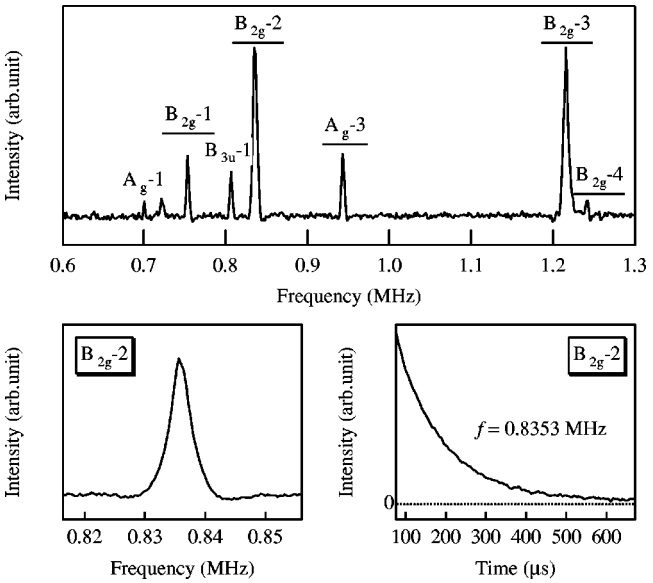

FIG. 1. EMAR resonance spectrum at room temperature (upper), the resonance peak of the B_{2g}-2 mode (lower left), and the free decay at this resonance frequency (lower right). Underlined modes were used for determination of the elastic constants.

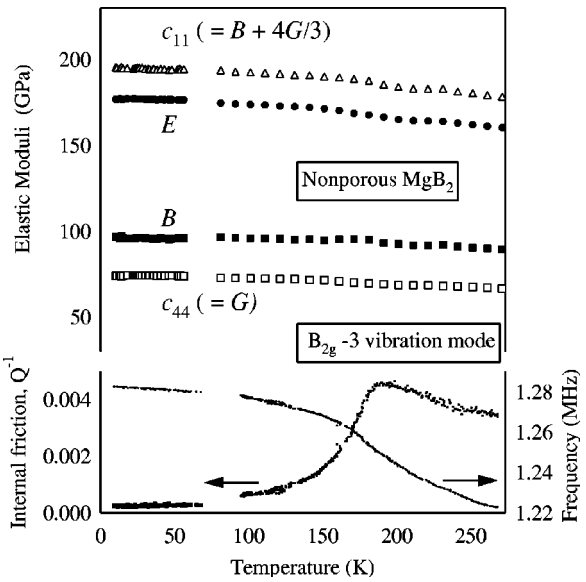

FIG. 2. Temperature dependence of elastic constants (upper) of the nonporous MgB_2, internal friction, and resonance frequency (lower).

spectra.[19] The resonance frequencies are first calculated by the Rayleigh-Ritz method, assuming the elastic constants and using the dimensions and mass density of the specimen. Then, attaining good agreement between the measured and calculated spectra after iterations, the assumed elastic constants can be regarded as the true values.

No texture was found from the three-directional longitudinal-wave velocities by pulse-echo measurements. We then supposed a polycrystalline MgB_2 to be elastically isotropic. The elastic constants of *porous* MgB_2 are determined to be $c_{11}^{(p)}=136$ and $c_{44}^{(p)}=51.7$ GPa (at room temperature). We confirmed the validity with two more specimens of different sizes and virtually the same porosity.

From the determined elastic constants of the porous MgB_2, we deduced those for the nonporous material through a micromechanics model.[21] We assume that (i) the matrix is elastically isotropic, and (ii) the pores (inclusions) are spherical. The elastic constants $\mathbf{C}_{mat}$ of the nonporous MgB_2 are obtained by solving Eqs. (1)–(3) self-consistently:

$$\mathbf{C}_{mat}=[\mathbf{I}-c_1\mathbf{A}]^{-1}(\mathbf{C}_{com}-c_1\mathbf{C}_{inc}\mathbf{A}), \tag{1}$$

$$\mathbf{A}=[c_0\mathbf{I}+c_1\mathbf{B}]^{-1}\mathbf{B}, \tag{2}$$

$$\mathbf{B}=[\mathbf{I}+\mathbf{S}\mathbf{C}_{mat}^{-1}(\mathbf{C}_{inc}-\mathbf{C}_{mat})]^{-1}. \tag{3}$$

Here, $\mathbf{C}_{com}$ are the measured elastic constants of the porous MgB_2, $\mathbf{C}_{inc}$ are those of the inclusion (now $\mathbf{C}_{inc}=\mathbf{0}$), $\mathbf{I}$ is the unit matrix, c_0 and c_1 are the volume fractions of the matrix and the pores, respectively, and $\mathbf{S}$ is the Eshelby tensor[22] for the spherical inclusion, which depends on $\mathbf{C}_{mat}$ and the pore shape.

Figure 2 shows the temperature dependence of the elastic constants, $c_{11}(=B+4G/3)$, $c_{44}(=G)$, Young's modulus E, and the bulk modulus B, of *nonporous* MgB_2 (upper part) and the normalized attenuation coefficient Q^{-1} of the B_{2g}-3 mode (lower part). In the behavior of the elastic constants, we found no anomaly such as lattice instability as seen in the $A15$ superconductors.[23] The polycrystalline elastic constants extrapolated to 0 K are $c_{11}=196$, $c_{44}=74.2$, $E=177$, and $B=96.6$ GPa. Our Young's modulus is close to 167 GPa (uncorrected for porosity, impurities, and temperature) acoustically measured by Cordero *et al.*,[17] and the longitudinal and transverse sound velocities at 77 K (Ref. 18) are very consistent with our measurements. However, the bulk modulus appears to be very different from the *isothermal* bulk modulus by x-ray measurements[16] and the *ab initio* calculations;[14,15] B ranges from 120 to 160 GPa in the literature.

TABLE I. Lattice properties and bulk moduli of AlB_2-type compounds at room temperature. $\sqrt{B/\rho}$ is a quantity with a dimension of the sound velocity.

	c/a	ρ (kg/m^3)	B (GPa)	$\sqrt{B/\rho}$ (m/s)	Reference
TiB_2	1.067	4491	243	7364	24
CrB_2	1.305	5225	233	6777	25
ZrB_2	1.112	6139	239	6239	25
MgB_2 [a]	1.142	2625	120-160	6761-7807	14–16
MgB_2	1.142	2625	86.1	5728	present work

[a]At low temperature (or 0 K).

According to the calculation,[15] the bulk and Young's moduli are 158 and 299 GPa for the isotropic MgB_2, respectively. Probably, the *ab initio* calculations are based on the elastic property of TiB_2, which has the AlB_2 crystallographic structure as MgB_2. Table I shows the lattice properties and the bulk moduli of TiB_2,[24] CrB_2, ZrB_2[25], and MgB_2.[14–16] The compound TiB_2 has the ideal axial ratio $c/a=1.067$ for the AlB_2 structure, i.e., a close-packed structure; therefore, the bond strength is predicted to be stronger than those of the other compounds. The present study indicates that the elastic property of MgB_2 is similar to that of ZrB_2 (rather than TiB_2) in terms of both axial ratio c/a and sound velocity $\sqrt{B/\rho}$. The elastic stiffness of MgB_2 is not particularly large among these compounds, as naturally predicted from its low condensation. From the above viewpoints, the present elastic constants are more reliable and reasonable.

The Debye temperature Θ_D is an important quantity to estimate the electron-phonon coupling constant λ, which is proportional to the mean sound velocity v_m,

$$\Theta_D=\left(\frac{h}{k_B}\sqrt[3]{\frac{3}{4\pi\Omega}}\right)v_m, \quad \frac{3}{v_m^3}=\frac{1}{v_l^3}+\frac{2}{v_s^3}, \tag{4}$$

where h/k_B has the usual meaning in quantum mechanics, Ω is the mean atomic volume, and v_l and v_s are the isotropic sound velocities given by $[(B+4G/3)/\rho]^{1/2}$ and $(G/\rho)^{1/2}$, respectively. The present elastic constants at 0 K yield $\Theta_D=819$ K. According to the McMillan formula,[26] the critical temperature T_c is expressed using Θ_D, λ, and the Coulomb pseudopotential μ^* as

$$T_c = \frac{\Theta_D}{1.45}\exp\left[\frac{-1.04(1+\lambda)}{\lambda-\mu^*(1+0.62\lambda)}\right]. \quad (5)$$

$T_c = 39$ K requires $\lambda \approx 0.76$–0.89 with the conventional values of $\mu^* \approx 0.05$–0.1; MgB_2 is thus within the intermediate-coupling regime. Nevertheless, the high Debye temperature (owing to the light component elements) contributes to its high T_c.

As seen in Fig. 2, a clear internal-friction peak appears at about 180 K, where the curvature of the resonance frequency changes and a slight modulus variation appears. A similar peak was first reported by Cordero *et al.*;[17] their peak temperature was located within 70–120 K, being different from our measurement, which is probably attributed to the difference in the frequency ranges used in both measurements (we used ~1 MHz, while they used 5–73 kHz). Thus, this large peak at about 180 K, showing a frequency dependence, can be a relaxation type like a Bordoni peak and not related to the superconductivity. In our measurement, since the large peak was located at relatively high temperature (180 K), the ultrasonic attenuation at the low-temperature region near T_c was possible to be studied, without being obscured by the large peak.

Figure 3 (upper part) shows the square of the resonance frequency and the internal friction of mode B_{2g}-2 near T_c. The square of frequency corresponds to the elastic constant representative for the resonance vibration. Obviously, the slope of the f^2 curve decreases below T_c, which indicates that the elastic softening occurs with the superconducting transition. However, the degree of softening is much less than in monoatomic superconductors[27] and the $A15$ superconductors.[23] Less softening means that the lattice still remains stiff below T_c, which results in the high Debye frequency.

It is noticeable that two very sharp attenuation peaks appear both in mode B_{2g}-2 and in mode B_{2g}-3 below 30 K, see Fig. 3. However, peak 2 is clearly shifted in temperature between B_{2g}-2 and B_{2g}-3. A third peak marked T_c appears in mode B_{2g}-2, but is absent in B_{2g}-3. These findings make it difficult to interpret our results at this stage. Currently we are investigating the possibility that the peaks are linked to the superconducting state. At the same time we have to exclude effects due to sample porosity.

In conclusion, we have examined the elastic property and

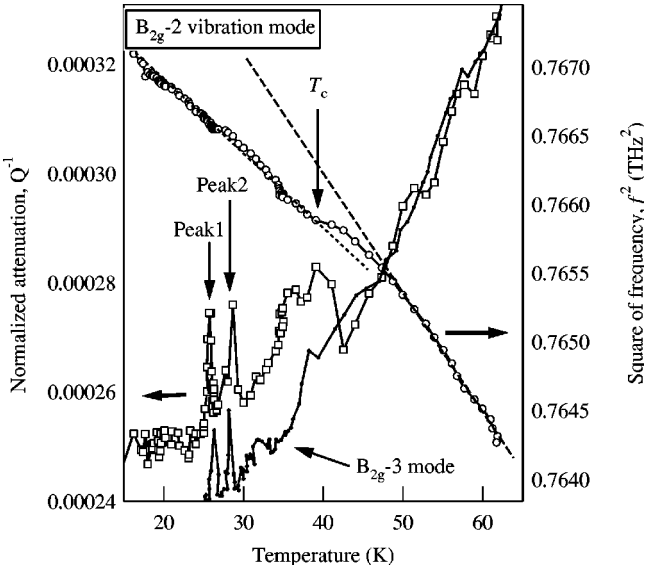

FIG. 3. Temperature behaviors of the square of resonance frequency and the internal friction of the B_{2g}-2 mode near T_c showing two peaks at 26 and 29 K. The dotted line shows Q^{-1} of the B_{2g}-3 mode, being shifted downward to avoid the overlap of the two curves. The background attenuation is $\sim 2.5\times10^{-4}$ μs/MHz. The magnetic field of ~0.2 T was applied for the EMAR excitation.

ultrasonic attenuation behavior of the high-T_c intermetallic superconductor MgB_2. The salient results are summarized as follows:

(i) The elastic constants are determined to be $B = 96.6$ and $G = 74.2$ GPa at 0 K through the micromechanics calculation. The bulk modulus B is smaller than the previous reports. However, the presented value is more reliable and reasonable in terms of both the axial ratio of the AlB_2 structure and its low condensation.

(ii) The elastic constants determined in this work yield the Debye temperature $\Theta_D = 819$ K and the intermediate coupling constant $\lambda \approx 0.76$–0.89 with the isotropic McMillan formula.

(iii) In the temperature behavior of ultrasonic attenuation, two anomalous peaks were observed.

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